Qualification of the Savannah River Site $^{252}\text{Cf}$ Shuffler for Receipt Verification Measurements of Mixed U-Pu Oxides

Franklin H. DuBose, PhD
Savannah River National Laboratory
April 20, 2012
In this talk…

Motivation
Overview of the K-Area $^{252}$Cf Shuffler
Principle of Operation
Calibration
Results
Motivation

K-Area work revolves around two programs

3013 Surveillance

Material Receipt/Storage
Both require extensive Pu measurement capabilities

Required similar $^{235}\text{U}$ measurement capabilities
• Mixed Oxide
• $\text{U}_3\text{O}_8$
• U-metal
$^{252}$Cf Shuffler

- 60 10-atm $^3$He Tubes
- 2 4-atm flux monitors
- $\sim$600 µg $^{252}$Cf source
- PLC
- JSR-15 Shift Register
- Derandomizer
- JSB-96
$^{252}\text{Cf} \text{ Shuffler Layout}$

- Flux Monitor
- High Density Polyethylene Moderator
- $^{252}\text{Cf}$ Source Storage
- Cadmium Liner
- $^{3}\text{He}$ Tubes
Neutron Counting

Source Irradiates at ~ $10^9$ n/s/g

Resulting in emitted neutrons that are:

Prompt: Immediate

Delayed: seconds – minutes after the initial irradiating event

Delayed neutrons are counted for several seconds after removing the source.
Delayed Neutron Production

$^{252}\text{Cf}$ Source

Prompt Neutrons

Fissile Material

β⁻ decay

γ emission

Delayed Neutrons

Stable Nuclei
Principle of Operation

\[ ^{252}\text{Cf} \]

Source

\[ ^{3}\text{He} \text{ Tubes} \]

Fissile Material
Calibration

Determined optimal assay parameters:

- ~ 1 s Forward time/Reverse Time
- 20 s Irradiation Time
- 10 s Delayed Neutron Count Time
- 70 Shuffles

Performed simulations of selected standards: MCNPX
Calibration Standards

High Purity

PuO$_2$ (5)
- 200 – 1300g
- 80 – 94% $^{239}$Pu

UPu (6)
- 0.1 – 1300g Pu
- 94% $^{239}$Pu
- 400 – 1700g U
- 93% enriched

Working Standards

• PuO$_2$ (7)
  - 400 – 4000g
  - 75 – 94 % $^{239}$Pu

• UPu (3)
  - 130 – 860g Pu
  - 88 – 94% $^{239}$Pu
  - 1600 – 4000g U
  - 80 - 94%
## Measurement and Simulation

<table>
<thead>
<tr>
<th>Measurement</th>
<th>Simulation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Each item measured five (3 - 5) times, with a 15 minute hold between measurements</td>
<td>MCNP5/MCNPX biased using time and energy dependent weight windows</td>
</tr>
<tr>
<td>Inner containers from HRS, LLNL, RFETS, LANL</td>
<td>Dimension of the HRS container used in all cases</td>
</tr>
<tr>
<td>Operating parameters identical for all items</td>
<td>Material density varied from 2.5 – 4.0 g/cc to accommodate 3013 volume</td>
</tr>
</tbody>
</table>
Plutonium Oxide Delayed Neutron Response

Measured PuO₂ Response

Plutonium Oxide Delayed Neutron Response

\[ DN(cps) = 0.669m_{239} + 166 \]
Simulated and Measured MOX Response

Delayed Neutron Count Rates
UPu Standards

Simulated and Measured MOX Response
## Measured vs Simulated DN Rates: UPu Standards

<table>
<thead>
<tr>
<th>Standard</th>
<th>Percent Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>-1.20</td>
</tr>
<tr>
<td>2</td>
<td>0.30</td>
</tr>
<tr>
<td>3</td>
<td>-0.73</td>
</tr>
<tr>
<td>4</td>
<td>0.17</td>
</tr>
<tr>
<td>5</td>
<td>2.35</td>
</tr>
<tr>
<td>6</td>
<td>-0.25</td>
</tr>
<tr>
<td>7</td>
<td>1.18</td>
</tr>
<tr>
<td>8</td>
<td>-3.49</td>
</tr>
<tr>
<td>9</td>
<td>1.06</td>
</tr>
</tbody>
</table>
The simulations showed that…

- delayed neutron contributions $^{235}\text{U}$ and $^{239}\text{Pu}$ dominated the response
- $^{239}\text{Pu}$ cps/g highly dependent on relative concentration
- average delayed neutron response behavior is viable for prediction
- density must be considered
Calibration Methodologies

Adjusted Count Rate Method

- Decreases the measured delayed neutron count rate by subtracting out the $^{239}\text{Pu}$ activation contribution
- Determines $^{235}\text{U}$ content from the $^{235}\text{U}$ cps/g
- Limited by $^{239}\text{Pu}$ content

Combined Count Rate Method

- Delayed neutron count rate is a linear combination of $^{235}\text{U}$ and $^{239}\text{Pu}$ activation contributions
- Coefficients depend on material density
$^{239}$Pu Ratio Dependent Delayed Neutron Count Rates

\[ \text{DN(cps/g)} = -0.173R + 0.672 \]
Adjusted Count Rate Method

\[ DN_{adj} = DN_{Net} - \left[ -0.1725 \ln(R) + 0.6716 \right] \cdot m_{239} \]

\[ m_{235} = \frac{DN_{adj}}{\left[ -0.0501 \ln(R) + 1.7602 \right]} \]
Combined Count Rate Method

\[ CR_{Net} = k_{235} m_{235} + k_{239} m_{239} \]

\[ m_{235} = \frac{DN_{Net} - k_{239} m_{239}}{k_{235}} \]

\[ k_{235} \approx \begin{cases} 2.0 \text{cps/g} & \text{, low density} \\ 1.4 \text{cps/g} & \text{, high density} \end{cases} \]
## Calibration Method Comparison

<table>
<thead>
<tr>
<th>Adjusted Count Rate</th>
<th>Combined Count Rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>High precision and accuracy over the applicable range</td>
<td>High precision and accuracy across the entire measured range</td>
</tr>
<tr>
<td>Under predicts $^{235}\text{U}$ content at higher masses</td>
<td>Used to successfully verify $\sim 94%$ of all UPu items measured to date</td>
</tr>
<tr>
<td>Less accurate with higher Pu content</td>
<td>Average Percent Difference: $&lt; 7.0%$</td>
</tr>
<tr>
<td>Has been successfully applied in the field on several items</td>
<td></td>
</tr>
</tbody>
</table>
Two Active calibration methods have been developed to quantify $^{235}\text{U}$ in mixed oxide

Method 1 is $^{239}\text{Pu}$ ratio dependent

Method 2 assumes independent contributions from each fissile nuclide

Both methods show high accuracy within applicable mass ranges

Method 1 and 2 have been successfully applied to verify over 100 mixed U-Pu bearing items
Planned and Completed Work

- Multiplicity characterization and measurement of total Pu in PuO$_2$ and Mixed Oxide – Complete
- Further assessment of $^{239}$Pu ratio dependencies - Complete
- Feasibility studies for “impure” metals and high Be oxides – Complete
- Feasibility studies for known-α analysis of oxides - Complete
- Qualification for verification of total Pu in pure and impure metals – In Progress
- Qualification for verification of total Pu in impure oxides – In Progress
- Qualification for verification of total U and Pu in mixed metals – Not Started